IN THE UNITED STATES PATENT AND TRADEMARK OFFICE
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

In re the Application of: **Toshio NARITA et al.** Group Art Unit: **1753**

Application Number: **10/501,813** Examiner: **Edna Wong**

Filed: **April 5, 2005** Confirmation Number: **6606**

For: **METHOD FOR FORMING Re ALLOY COATING FILM HAVING HIGH Re CONTENT THROUGH ELECTROPLATING**

Attorney Docket Number: **042393**
Customer Number: **38834**

SUBMISSION OF APPEAL BRIEF

Commissioner for Patents
P.O. Box 1450
Alexandria, Virginia 22313-1450

November 5, 2007

Sir:

Appellant submit herewith an Appeal Brief in the above-identified U.S. patent application.

Attached please find a check in the amount of $510.00 to cover the cost for the Appeal Brief.

If any additional fees are due in connection with this submission, please charge our Deposit Account No. 50-2866.

Respectfully submitted,

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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

APPEAL BRIEF FOR THE APPELLANT

Ex parte **Toshio NARITA et al.** (Appellant)

METHOD FOR FORMING Re ALLOY COATING FILM HAVING HIGH Re CONTENT THROUGH ELECTROPLATING

Serial Number: **10/501,813**

Filed: **April 15, 2005**

Appeal No.:

Group Art Unit: **1753**

Examiner: **Edna Wong**

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Date: **November 5, 2007**

BRIEF ON APPEAL

(I) REAL PARTY IN INTEREST

The real party in interest is **JAPAN SCIENCE AND TECHNOLOGY AGENCY, EBARA CORPORATION,** and **SAPPORO ELECTROPLATING INDUSTRIAL CO., LTD.** by an assignment recorded in the U. S. Patent and Trademark Office on **April 7, 2005** at Reel **016429**, Frame **0665**.

(II) RELATED APPEALS AND INTERFERENCES

There are no other appeals or interferences known to appellant, appellant's legal representative, or assignee that will directly affect or be directly affected by or have a bearing on the Board's decision in the pending appeal.

(III) STATUS OF CLAIMS

Claims 1, 2 and 6 are pending and are appealed.

(IV) STATUS OF AMENDMENTS

No amendments have been filed subsequent to the close of prosecution.

(V) SUMMARY OF CLAIMED SUBJECT MATTER

The present invention relates to a method for forming a high-Re-content alloy film which is used as a corrosion-resistant alloy coating for high-temperature components or the like (page 1, lines 4-5).

A Ni-based superalloy substrate is required to have high oxidation resistance and corrosion resistance. Such required high-temperature oxidation resistance has been obtained through a surface diffusion treatment. Rhenium (Re) can be used as the diffusion barrier layer to provide enhanced high-temperature corrosion resistance. Re, having excellent thermal shock resistance, is also used as high-temperature members or components of various combustors, such as a rocket-engine combustor, or high-temperature nozzles (page 1, lines 10-20).

There are Re alloy films such as a Ni-Cr-Re alloy film having a Re content of up to 50 weight% (this percentage becomes lower when converted into atomic composition ratio), a Ni-Co-Re alloy film, and a Re-Ni alloy film for electric contacts, which has a Re content of up to 85 weight% (63 atomic%). In all of the above plated films, the content of Re is in a low level (page 2, lines 1-7).

An object of the present invention is to provide a method capable of forming a Re alloy film on a surface having a complicated shape, which cannot be achieved by a sputtering process or physical deposition process. Another object of the present invention is to provide a method capable of forming a Re alloy film at a thin thickness, which cannot be achieved by a thermal spraying process. Still another object of the present invention is to provide a method of forming a higher-Re-content alloy film through an electroplating process at a low cost in a

simplified manner as compared to the physical deposition process and the thermal spraying process (page 2, lines 10-17).

Through various researches on a Re electroplating process, the inventors found that a Re alloy film having a Re content of 98% or more by atomic composition can be formed by adding into an electroplating bath an organic acid having at least one functional group selected from the group consisting of a hydroxyl group, a carbonyl group and an amino group, and controlling the respective compositions of the organic acid and a metal ion in the electroplating bath (page 2, lines 18-23).

Specifically, the present invention is a method for forming a Re alloy film through an electroplating process using an electroplating bath containing an aqueous solution. In this method, the aqueous solution includes a perrhenate ion in a concentration of 0.1 to 8.0 mol/L, at least one ion selected from the group consisting of nickel, iron, cobalt and chromium (III) ions, in a total concentration of 0.005 to 2.0 mol/L, at least one of a lithium ion and a sodium ion, in a total concentration of 0.0001 to 5.0 mol/L, and at least one organic acid selected from the group consisting of carboxylic acid, hydroxyl-carboxylic acid and amino acid, in a concentration of greater than 5.0 to 15.0 equivalents to the concentration of all of the metal ions. Further, the electroplating bath has a pH of 0 to 8, and a temperature of 10 to 80°C. In this manner, this method can form a high-Re-content alloy film which contains Re at 98 % or more by atomic composition, and serves as a heat/corrosion resistant alloy coating (page 2, line 24 to page 3, line-6).

In the method of the present invention, the alloy film to be formed may have a composition consisting of 98% or more, by atomic composition, of Re, with the remainder being at least one selected from the group consisting of Ni, Co, Fe, Mn, Cr, Mo, W, Nb, Ta, Hf, Si, Al, Ti, Mg, Pt, Ir, Rh, Au, Ag, P, B, C, Y and Ce, and inevitable impurities. This allows desired functions to be given to the film depending on a substrate and an intended purpose (page 3, lines 7-12).

In the method of the present invention, if the concentration of perrhenate ion is less than 0.1 mol/L, a resulting plated film cannot contain Re at 98% or more. Further, the concentration of perrhenate ion greater than 8.0 mol/L causes creation of an insoluble substance in the bath. As to the at least one ion selected from the group consisting of nickel, iron, cobalt and chromium (III) ions, the use of a total concentration of the ion less than 0.005 mol/L causes significant deterioration in plating efficiency. Further, if the total concentration of the ion is greater than 2.0 mol/L, the Re content in a resulting plated film will be less than 98% by atomic composition. **As to the at least one of a lithium ion and a sodium ion, if the total concentration of the ion is less than 0.0001 mol/L, the Re content in a resulting plated film will be less than 98% by atomic composition**. Further, the use of a concentration of the ion greater than 5.0 mol/L causes creation of an insoluble substance in the bath to result in deteriorated flowability of the aqueous solution (page 3, lines 13-25).

If the concentration of the at least one organic acid selected from the group consisting of carboxylic acid, hydroxycarboxylic acid and amino acid, is equal to or less than 5.0 equivalents to the concentration of all of the metal ions, the Re content in a resulting plated film will be less

than 98% by atomic composition. Further, the use of a concentration of the at least one organic acid greater than 15.0 equivalents causes creation of an insoluble substance in the bath to result in deteriorated flowability of the aqueous solution (page 3, line 26 to page 4, line 2).

(VI) GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

Whether claims 1, 2 and 6 are obvious over Castonguay (U.S. Patent No. 3,857,683) and Phillips (U.S. Patent No. 3,704,211) under 35 USC §103(a).

(VII) ARGUMENT

Claims 1, 2 and 6 stand rejected under 35 USC §103(a) as being obvious over Castonguay (U.S. Patent No. 3,857,683) and Phillips (U.S. Patent No. 3,704,211)

First, it should be noted that the present invention is directed to "a method for forming a high-Re-content alloy film which contains Re at 98 % or more by atomic composition." Nothing in Castonguay and Phillips teaches or suggests how such a high-Re-content alloy film is obtained.

Through extensive researches on a Re electroplating process, the inventors discovered that a Re alloy film having a Re content of 98% or more by atomic composition can be formed by an electroplating bath containing an organic acid having at least one functional group selected from the group consisting of a hydroxyl group, a carbonyl group and an amino group, and controlling the respective compositions of the organic acid and a metal ion in the electroplating

bath. At the same time, the interrelated conditions of the electroplating bath have to be satisfied to obtain a Re alloy film having a Re content of 98% or more by atomic composition.

As explained in the specification at page 3, if the concentration of perrhenate ion is less than 0.1 mol/L, a resulting plated film cannot contain Re at 98% or more. On the other hand, the use of a concentration of perrhenate ion greater than 8.0 mol/L causes creation of an insoluble substance in the bath. As to the at least one ion selected from the group consisting of nickel, iron, cobalt and chromium (III) ions, the use of a total concentration of the ion less than 0.005 mol/L causes significant deterioration in plating efficiency. Further, if the total concentration of the ion is greater than 2.0 mol/L, the Re content in a resulting plated film will be less than 98% by atomic composition.

Also, **if the total concentration of a lithium ion and a sodium ion is less than 0.0001 mol/L, the Re content in a resulting plated film will be less than 98% by atomic composition**. On the other hand, the use of a concentration of the ion greater than 5.0 mol/L causes creation of an insoluble substance in the bath to result in deteriorated flowability of the aqueous solution.

Even where these conditions are satisfied, if the concentration of the at least one organic acid selected from the group consisting of carboxylic acid, hydroxycarboxylic acid and amino acid, is equal to or less than 5.0 equivalents to the concentration of all of the metal ions, the Re content in a resulting plated film will be less than 98% by atomic composition. On the other hand, the use of a concentration of the at least one organic acid greater than 15.0 equivalents

causes creation of an insoluble substance in the bath to result in deteriorated flowability of the aqueous solution.

Thus, claim 1 specifies the interrelated conditions of the electroplating bath for forming a high-Re-content alloy film which contains Re at 98 % or more by atomic composition. The combination of Castonguay and Phillips does not teach or suggest the claimed invention. Also, it would not have been predictable that the claimed method can produce a high-Re-content alloy containing Re at 98 % or more.

A. Castonguay Does Not Teach Or Suggest How A High-Re-Content Alloy Film Which Contains Re At 98 Atomic % Is Obtained.

The Examiner Admits that Castonguay does not disclose (a) "at least one of a Li ion and a Na ion, in a total concentration of 0.0001 to 5.0 mol/L" as recited in claim 1. The Examiner admits that Castonguay does not disclose (b) "said aqueous solution further includes at least one ion selected from the group consisting of potassium, rubidium, cesium, calcium, strontium and barium ions, wherein the total concentration of said at least one of lithium ion and sodium ion in said electroplating bath is greater than the total concentration of said at least one ion selected from the group consisting of potassium, rubidium, cesium, calcium, strontium and barium ions." as recited in claim 6. (Office Action mailed 05/18/07, page 4).

Regarding the importance of Li ion and Na ion, the present specification specifically explains that "if the total concentration of a lithium ion and a sodium ion is less than 0.0001

mol/L, the Re content in a resulting plated film will be less than 98% by atomic composition" (present specification page 3, lines 20-25).

The Examiner alleged as follows: "This method step is taught by the combination of Castonguay and Phillips, e.g., 4.5-5.5 g/l Na (= 0.20 to 0.24 mol/l Na) [Phillips, col. 4, line 43] in the bath of Example XXIII (Castonguay, col. 9, lines 21-36)." (Advisory Action mailed 8/14/07, at page 3).

Castonguay explains as follows:

> Briefly, this invention comprehends a **novel printed circuit board material** in the form of a layered stock comprising an insulating support, at least one layer of electrical resistance material adhering to said support, and a layer of a highly conductive material adhering to the resistance material and in intimate contact therewith, **said layer of electrical resistance material** being selected from the group consisting of chromium-antimony, chromium-manganese, chromium-phosphorus, chromium-selenium, chromium-tellurium, cobalt-antimony, cobalt-boron, cobalt-germanium cobalt-indium, cobalt- molybdenum, cobalt-phosphorus, **cobalt-rhenium**, cobalt-ruthenium, cobalt-tungsten, cobalt-vanadium, iron-vanadium, nickel-antimony, nickel- boron, nickel-chromium, nickel-germanium, nickel-indium, nickel- molybdenum, nickel-phosphorus, **nickel-rhenium**, nickel-vanadium and palladium-molybdenum.

(Castonguay, column 1, lines 29-46, emphasis added). Thus, Castonguay discloses **printed circuit board materials** and discusses cobalt-rhenium system and nickel-rhenium system as the **electrical resistance materials**. However, Castonguay does not teach or suggest that a high-Re-content alloy film which contains Re at 98 atomic % is obtained. Castonguay describes, at the portion referred to by the Examiner, as follows:

EXAMPLE XXIII

System: Cobalt-Rhenium
Composition: 25 to 95% rhenium
Resistivity: 135 to 438 microhm-cm
TCR: plus 300 to plus 100 ppm/°C
Plating Techniques

Potassium perrhenate, $KReO_4$	1-150	g/l
Cobalt (sulfate), Co^{+2} (SO_4^{-2})	2-25	g/l
Citric Acid, $HOC_3H_4(COOH)_3$	20-200	g/l
Current density	2-12	amp/dm²
Temperature	25-90	°C
pH (by addition of ammonia NH_4OH; or sulfuric acid H_2SO_4)	3-8	

By varing the rhenium content in the bath from 3 to 80%, the rhenium content in the deposit may be varied from 25 to 95%.

(Castonguay, column 9, lines 21-36). Thus, Castonguay describes: "By varying the rhenium content in the bath from 3 to 80%, **the rhenium content in the deposit may be varied from 25 to 95 %.**" In Castonguay, the deposit contains rhenium 95 weight % at the most. This indicates that high-Re-content alloy containing Re at 98 atomic % or more is not obtained by Castonguay. The upper limit of Re content 95 weight % is converted to atomic percentage as follows:

$$95 \text{ weight \%} = \frac{95/186.2}{(95/186.2)+(5/58.9)} \text{ atomic \%} = 85.8 \text{ atomic \%},$$

where 186.2 is the atomic weight of Re and 58.9 is the atomic weight of Co.

Thus, the value 95 weight % amounts to 86 atomic % or less. Thus, Castonguay does not disclose how a high-Re-content alloy containing Re at 98% or more by atomic composition is

obtained. The same thing can be said about nickel-rhenium system because the atomic weight of Ni is 58.7, which is about the same as that of Co.

As explained in the present specification, for example, at page 3, if the total concentration of a lithium ion and a sodium ion is less than 0.0001 mol/L, the Re content in a resulting plated film will be less than 98% by atomic composition.

B. Phillips Is Irrelevant To Obtaining High-Re-Content Alloy Film.

The Office Action cites Phillips in an attempt to remedy the above deficiencies of Castonguay. Phillips discloses a method for electroplating **Ni-Co ferromagnetic films** and an improved electroplating for the method, particularly, the electroplating of nickel-cobalt or cobalt magnetic films for use in the field of recording. **Rhenium is added to serve as a grain refiner, usually up to approximately 1% of the final composition by weight** (Phillips, col. 3, lines 50-54). Thus, according to Phillips, the obtained film is Ni-Co ferromagnetic film contains rhenium at most as low as 1 weight %.

Therefore, Phillips is irrelevant to obtaining a high-Re-content alloy film. There is no reason for a person of ordinary skill in the art to combine the two references.

More specifically, Phillips discloses a process for depositing a nickel-cobalt magnetic electroplate. Regarding addition of rhenium, Phillips describes as follows:

> It has been discovered that the **addition of rhenium to a nickel-cobalt or a cobalt electroplating bath** in a small but effective amount appears to serve as a grain refiner, usually **up to approximately 1% of the final**

> **composition by weight** and has the unexpected result of modifying and allowing control of the *Mr/Hc* ratio of the deposited film.

(Phillips, col. 3, lines 50-55). Thus, Re is added as a grain refiner and to modify or control *Mr/Hc* ratio. Even if Philips teaches use of Na ion in such a nickel-cobalt or a cobalt electroplating bath, it teaches nothing about high Re-content alloy. The final composition contains rhenium as low as 1 weight % or less. Phillips describes, at the portion referred to by the Examiner, as follows:

TABLE I

[Grams/liter]

Type of ion in solution	General range	Preferred range	Preferred embodiment
Ni	0-26	22-26	26
Co	10-13	10-12	12
Re	.06-1.3	.13-.16	.32
P	0-0.7	.01-.14	0
Na	4.5-5.5	4.5-5.5	5
NH_4	7-10	7-8	8
pH	3-5	4-4.5	4.5
Temperature (° C.)	20-55	21-24	21-24
Current density, amp/sq. ft.	.1-30	10-20	10

The Examiner appears to allege that "4.5-5.5 g/l Na" (=0.20 to 0.24 mol/l Na) in the above disclosure in Phillips make it obvious to add the similar amount of Na in the electroplating bath of Example XXIII disclosed in Castonguay. However, Castonguay discusses a printed circuit board material while Phillips discusses electroplating of magnetic films for high-density recording. Thus, the uses of the obtained films are different between Castonguay and Phillips. Also, the compositions of the obtained films are different between Castonguay and Phillips.

Therefore, a person of ordinary skill would not think of combining the disclosures of Castonguay and Phillips. The Office Action does not specify the reason why a person of ordinary skill in the art would apply the content of sodium in Phillips to the electroplating bath of Castonguay. The Examiner is required to specifically explain the reason to combine the known elements in the fashion claimed. KSR International Co. v. Teleflex Inc., 127 S. Ct. 1727, 1741 (April 30, 2007) (the Supreme Court ruled that the proper question for evaluating obviousness is "whether there was an apparent reason to combine the known elements in the fashion claimed").

Moreover, the embodiments in the present specification demonstrate unexpected results that high-Re-content alloy films which contain Re at 98 % or more by atomic composition are obtained. Neither Castonguay nor Phillips recognized how a high-Re-content alloy film which contains Re at 98 % or more by atomic composition can be obtained.

C. Allegations In The Office Action Do Not Support The Obviousness Of Claimed Invention Over Castonguay And Phillips.

The Examiner alleged in Advisory Action at page 3 as follows:

> This method step does not distinguish the method over the prior art because:
> (a) Similar processes can reasonably be expected to yield products which inherently have the same properties. *In re Spade* 15 USPQ 2d 1655 (CAFC 1990); *In re DeBlauwe* 222 USPQ 191; *In re Wiegand* 86 USPQ 155 (CCPA 195);
> (b) If the composition is physically the same, it must have the same properties; . . . .

It may be true that, if the composition is physically the same, it must have the same properties. However, neither Castonguay nor Phillips discloses physically the same composition. That is why Office Action does not allege that Castonguay or Phillips anticipates the claimed invention.

Also, although the Examiner alleges that these prior art references are expected to yield products which inherently have the same properties, Castonguay describes that the rhenium content may be varied up to 95 wt % (85.8 atomic %), and nothing in Castonguay indicates that a high-Re-content alloy containing Re of 98 atomic % or more is obtained. The Examiner's allegation is not properly based on the actual disclosure in the reference.

The Advisory Action further alleged at page 3 as follows:

> (c) A preamble is not necessarily accorded any patentable weight where it merely recites the purpose of a process or the intended use of a structure, and where the body of the claim does not depend on the preamble for completeness but, instead, the process steps or structural limitations are able to stand alone. *In re Hirao* 535 F. 2d 67, 190 USPQ 15 (CCPA 1976) and *Kropa v. Robie* 187 F 2d 150, 152, 88 USPQ 478, 481 (CCPA 1951). . . .

However, there are differences between the presently claimed method and that disclosed in Castonguay or Phillips. There is no overlapping in the composition with Castonguay or Phillips. Also, Castonguay and Phillips do not disclose the specific interrelationship between the recitations. Thus, claim 1 patentably distinguishes regardless of the recitation of the preamble.

The Advisory Action further alleged at page 4 as follows:

> (d) The reason or motivation to modify the reference may often suggest what the inventor has done, but for a different purpose or to solve a different problem. It is not necessary that the prior art suggest the combination to achieve the same advantage or result discovered by the Applicants. *In re Linter* 458 F.2d 1013? 173 USPQ 560 (CCPA 1972); *In re Dillon* 919 F.2d 688, 16 USPQ2d 1897 (Fed. Cir. 1990), cert. denied, 500 US 904 (1991); and MPEP § 2144. . . . .

Even if it is not necessary that the prior art suggest the combination to achieve the same advantage or result discovered by the Applicants, it *is* required to identify the reason why a person of ordinary skill in the art would modify or combine the prior art to reach the claimed subject matter. The Supreme Court addressed the standard for obviousness in KSR International Co. v. Teleflex Inc., 127 S. Ct. 1727 (April 30, 2007). The Court explained that the proper question for evaluating obviousness is "whether there was an apparent reason to combine the known elements in the fashion claimed." KSR, at 1741 (slip op. at 14). Here, the record evidences no such reason.

The Advisory Action further alleged at page 4 as follows:

> (e) The claims attempt to define the subject matter in terms of the result to be achieved, which merely amounts to a statement of the underlying problem, without providing the technical method features necessary for achieving this result.

However, claim 1 does not try to define the subject matter in terms of the result to be achieved, but specifically recites a step of performing an electroplating process

using an electroplating bath which contain a particular aqueous solution which patentably distinguishes over the prior art.

For at least these reasons, claim 1 patentably distinguishes over Castonguay and Phillips. Claims 2 and 6, depending from claim 1, also patentably distinguish over Castonguay and Phillips for at least the same reasons.

CONCLUSION

For at least the foregoing reasons, the Examiner erred in finding that claims 1, 2, and 6 are obvious over the prior art. The Honorable Board is respectfully requested to reverse the rejection of the Examiner.

If this paper is not timely filed, Appellant hereby petition for an appropriate extension of time. The fee for any such extension may be charged to our Deposit Account No. 50-2866, along with any other additional fees that may be required with respect to this paper.

Respectfully submitted,

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(VIII) CLAIMS APPENDIX

1. (Rejected) A method for forming a high-Re-content alloy film which contains Re at 98 % or more by atomic composition, said method comprising performing an electroplating process using an electroplating bath which contains an aqueous solution including:

a perrhenate ion in a concentration of 0.1 to 8.0 mol/L;

at least one ion selected from the group consisting of nickel, iron, cobalt and chromium (III) ions, in a total concentration of 0.005 to 2.0 mol/L;

at least one of a Li ion and a Na ion, in a total concentration of 0.0001 to 5.0 mol/L; and

at least one organic acid selected from the group consisting of carboxylic acid, hydroxycarboxylic acid and amino acid, in a concentration of greater than 5.0 to 15.0 equivalents to the concentration of all of said metal ions,

wherein said electroplating bath has a pH of 0 to 8, and a temperature of 10 to 80°C.

2. (Rejected) The method as defined in claim 1, wherein said alloy film to be formed has a composition consisting of 98% or more, by atomic composition, of Re, with the remainder being at least one selected from the group consisting of Ni, Co, Fe, Mn, Cr, Mo, W, Nb, Ta, Hf, Si, Al, Ti, Mg, Pt, Ir, Rh, Au, Ag, P, B, C, Y and Ce, and inevitable impurities.

3-5. (Cancelled)

6. (Rejected) The method as defined in either one of claims 1 and 2, wherein said aqueous solution further includes at least one ion selected from the group consisting of potassium, rubidium, cesium, calcium, strontium and barium ions, wherein the total concentration of said at least one of lithium ion and sodium ion in said electroplating bath is greater than the total concentration of said at least one ion selected from the group consisting of potassium, rubidium, cesium, calcium, strontium and barium ions.

7. (Cancelled)

(IX) EVIDENCE APPENDIX

None Presented.

(X) RELATED PROCEEDINGS APPENDIX

No Related Proceedings